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## MAGNETIC BEHAVIOR AT THE IRON SITES IN Pd Fe Mn ALLOYS\*

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Abstract

Mossbauer Effect hyperfine measurements of  $(\text{Pd}_{0.9965}\text{Fe}_{0.0035})_{1-x}\text{Mn}_x$  have been obtained for  $1.1 < T < 30$  K and  $0 < H < 6T$  for  $x = 0.01, 0.05, 0.06, \text{ and } 0.065$ . Ferromagnetic ordering is clear for  $x = 0.01$  and is suggested for  $x = 0.05$  at 4 K. At 1.1 K ordering at the iron sites in all samples, except  $x = 0.01$ , is consistent with a spin-glass or antiferromagnetic phase.

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Running Title: Magnetic Behavior at the Iron Sites in Pd Fe Mn

For the system  $(\text{Pd}_{0.9965}\text{Fe}_{0.0035})_{1-x}\text{Mn}_x$  Verbaek et al. [1] have suggested three distinct regimes of magnetic behavior ranging from giant-moment ferromagnetism ( $x = 0.01$ ) to spin-glass ordering ( $x = 0.065$ ) with an intermediate region ( $x = 0.05$ ) in which a ferromagnetic phase becomes spin-glass at a lower temperature. These conclusions were based primarily on low field ac susceptibility measurements interpreted using the theory of Sherrington and Kirkpatrick. [2]

To supplement these and other bulk measurements on this system we have doped samples having  $x = 0.01, 0.05, 0.06, \text{ and } 0.065$  with  $^{57}\text{Co}$ , in order to study the internal local interactions at the iron sites using the Mossbauer Effect (ME) of  $^{57}\text{Fe}$ . The ME has often been used to detect the onset of the spin-glass freezing temperature. [3,4]

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The samples were arc cast from high purity elements. Losses during preparation, homogenization, and annealing were negligible so no chemical analysis was made. After the buttons were rolled to about 0.4-mm thickness, disks about 9-mm diam were cut and one of each was electroplated with about 1  $\mu$ Ci of  $^{57}\text{Co}$ , the parent of  $^{57}\text{Fe}$ . After a 2 - 18 h high-vacuum, diffusion-anneal at about 1050°C, the local concentration of  $^{57}\text{Co}$  was <1000 ppm. For a given x, ac susceptibility measurements of both doped and undoped samples confirmed the results of Verbeek et al.[1]. The Curie-Weiss temperatures  $\theta$  determined from the paramagnetic data below 24 K are given in Table 1. As in[1] we also present the spin-glass freezing temperature  $T_{\text{sg}}$  determined from c.v. susceptibility measurements.

The ME at the  $^{57}\text{Co}$  (iron) sites was determined as a function of temperature (1.1-30 K) and applied field (0-6 T) using a room temperature unsplit absorber and conventional ME spectrometer. The low temperature  $^{57}\text{Fe}$  isomer shift for each sample was the same ( $\pm 0.006$  mm/sec) as found in (pure) palladium. In the presence of a magnetic hyperfine field, splitting of the energy levels of  $^{57}\text{Fe}$  give rise to a six-line ME spectrum having areal intensities 3:1:1:1:1:3. The normalized intensity of the "2 - 5" lines I depends on the polarization or average direction of the effective field  $H_{\text{eff}}$  relative to the direction of the observed ME  $\gamma$ -rays; I varies from 0 (field parallel) to 4 (field perpendicular) and is 2 for random polarization. Analysis of resolved ME spectra in an applied field gives information about the nature of the magnetic order present.

We first examined the region above  $\theta$ , where a single ME line was found. In sufficient fields, applied perpendicular to the sample plane and parallel to the  $\gamma$ -ray analysis direction, four-line ME spectra ( $I = 0$ ) were observed. The splitting is proportional to  $H_{\text{eff}}$ , which is the vector sum of the applied field H and the internal field  $H_{\text{int}}$  acting on the  $^{57}\text{Fe}$  nucleus.

$H_{\text{int}}/H_{\text{sat}}$ , where  $H_{\text{sat}}$  is the saturation value of  $H_{\text{int}}$ , is taken to be equal to the relative local magnetization of the samples at a given x, T, and H.

$H_{\text{int}}/H_{\text{sat}}$  data were fitted to a modified Brillouin curve using  $H/(T - \theta_M)$  as the argument.  $\theta_M$  agrees fairly well with  $\theta$  (or  $T_{\text{sg}}$  for x = 0.065), as shown in Table 1. Aside from the  $\theta_M$  values, the other parameters determined from these Brillouin fits were the same for all four alloys, i.e.,  $H_{\text{sat}} = -28 \pm 0.8$  T and a moment  $\mu = 36 \pm 1 \mu_B$  for g = 2 and for T > 20. Iron in palladium gives  $-30.1 \pm 0.1$  T and  $12.1 \pm 0.3 \mu_B$ [5] if we recalculate  $\mu$  using g = 2.

Spontaneous ordering for each  $x$  was determined from the  $H = 0$  ME spectra at  $T < 2\theta$ . As the spontaneous ordering temperature  $T_M$  was approached from  $T > T_M$ , rapidly increased broadening was observed. For each  $x$  at  $T \ll T_M$ , the well resolved six-line ME spectrum we observed was taken as evidence that the  $^{57}\text{Fe}$  atoms were all in the same electronic state. As  $T$  was then raised  $H_{\text{int}}$  decreased, but the outer components of the ME spectra were progressively broadened. This overall behavior is consistent with a distribution of local environments about the  $^{57}\text{Fe}$  probe sites in these random alloys, each with its own  $T_M$ . We have fitted the ME spectra with a 2-site model to account partially for this effect. For each  $x$  we have fixed the relative proportions to be the binominal probability distribution function for zero or one nearest neighbor manganese atoms to the  $^{57}\text{Fe}$  sites. The  $H_{\text{int}}$  values determined for one- or two-site fits were used to determine experimental  $T_M$  values using a molecular-field description of the ordered state. For all  $x$ , a value of  $H_{\text{sat}} = -30.1 \pm 0.4$  T was obtained from the ordered data and is deemed more accurate than that obtained by extrapolating the paramagnetic data.  $T_M$  values, given in Table 1, are 10 - 20% higher than the  $\theta_M$  values, but this is not surprising in view of the difficulties in analyzing the data, the inadequacy of the models, and the likelihood that the transition temperature is not sharply defined.

In the absence of an applied field the temperature dependence of the magnetic behavior at the  $^{57}\text{Fe}$  sites in the region of spontaneous ordering was similar for all  $x$ ; no change was expected or found in the ME spectra that could be attributed to a spin-glass phase.

In an effort to distinguish between ferromagnetic and spin-glass behavior, we have studied the response of  $^{57}\text{Fe}$  in these alloys to an applied field at  $T \ll T_M$ . As discussed earlier, changes in the polarization of the spins imply changes in the ME spectra. From ME spectra taken near 1.1 K as a function of  $H$ , we have determined  $I$  and plotted these results in Fig. 1. For  $x = 0.01$  at  $H = 0$ ,  $I$  was 2.6 indicating a tendency toward spontaneous polarization in the sample plane. For  $H = 75$  mT,  $I$  was 0.1 indicating nearly complete polarization along the axis of  $H$ ; such easy alignment is characteristic of a ferromagnet. In contrast to this response, the  $x = 0.065$  sample was very difficult to polarize, as shown by the slow decrease of  $I$  with  $H$ . The well known spin-glass  $\text{Au}(\text{Fe})$  [4] and certain antiferromagnetic materials show a similar ME response to  $H$ . For  $x = 0.05$ , the composition that is claimed [1] to exhibit a ferromagnetic transition followed by a spin glass transition (Table 1), the

ME data at 1.1 K show that polarization is somewhat easier than for  $x = 0.065$  but quite unlike the ferromagnetic response of  $x = 0.01$ . The polarization response at 4.0 K for  $x = 0.05$  is shown in Fig. 1; this response is more like that of a ferromagnet, but some of this same trend is found for the  $x = 0.065$  sample. Moreover,  $H_{int}$  tends to -30.1 T (the fully ordered value) with increasing  $H$  for  $x = 0.05$  whereas  $H_{int}$  (assuming antiparallel alignment) tends to -31 T at  $H = 6$  T for the  $x = 0.065$  sample, indicating only partial alignment at the iron sites and providing secondary support that the ordering at 4 K is ferromagnetic for  $x = 0.05$ .

In summary,  $^{57}\text{Fe}$  ME data in these Pd(Fe Mn) alloys yield magnetic ordering temperatures comparable to those obtained by bulk measurements. Polarization data clearly show ferromagnetism for  $x = 0.01$  and are consistent with a spin-glass (or antiferromagnetic) phase at 1.1 K in the other alloys. The ME data do not fully support ordinary ferromagnetism at 4 K in the  $x = 0.05$  alloy.

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#### REFERENCES

- [1] B. H. Verbeek, G. J. Nieuwenhuys, H. Stocker, and J. A. Mydosh, Phys. Rev. Lett. 40 (1978) 586.
- [2] D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. 35 (1975) 1792.
- [3] J. A. Mydosh, J. Magn. Magn. Mater. 7 (1978) 237 and references therein.
- [4] B. Window, J. Magn. Magn. Mater. 1 (1975) 167.
- [5] M. P. Maley, R. D. Taylor, and J. L. Thompson, J. Appl. Phys. 38 (1967) 1249.

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Table 1. Transition temperatures for  $(\text{Pd Fe})_{1-x} \text{Mn}_x$ .

x	$T_{\text{sg}}$ (K)	$\theta$ (K)	$\theta_M$ (K)	$T_M^a$ (K)
0.01	—	15.2 (2)	14.1 (1)	19.2 (6)
0.05	2.5 (2)	10.0 (4)	10.7 (1)	12.9 (1) 12.2 (5)
0.06	5.0 (1)	6.5 (1)	5.8 (1)	7.7 (2) 6.2 (2)
0.065	5.1 (1)	3.5 (3)	6.1 (1)	7.0 (1) 6.4 (1)

<sup>a</sup>Double entries indicate 2-site fits.

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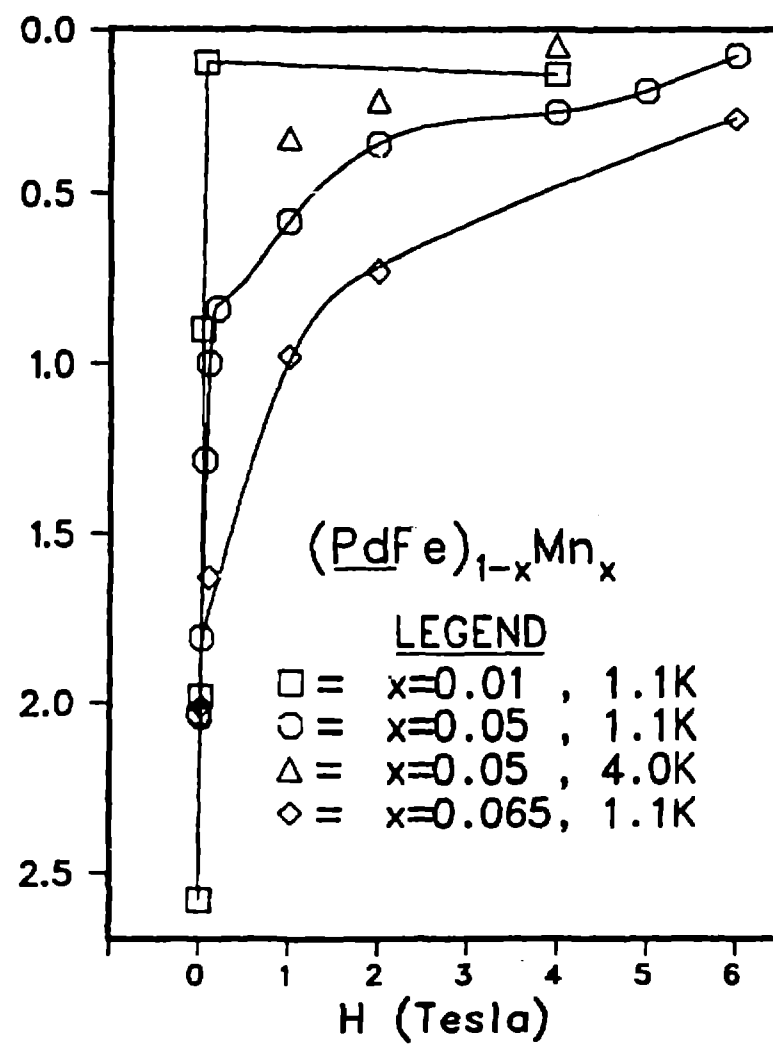


Fig. 1. Relative Line Intensity  $I$  versus  $H$  of  $(\text{PdFe})_{1-x}\text{Mn}_x$  for  $x = 0.01, 0.05$ , and  $0.065$  at  $1.1\text{ K}$  and  $x = 0.05$  at  $4\text{ K}$ .